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LLNL-JRNL-409505

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December 18, 2008

Sensor Letters

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Colossal Magnetoresistive Manganite Based Fast Bolometric X-ray Sensors for Total Energy Measurements of Free Electron Lasers

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ABSTRACT

Bolometric detectors based on epitaxial thin films of rare earth perovskite manganites have been proposed as total energy monitors for X-ray pulses at the Linac Coherent Light Source free electron laser. We demonstrate such a detector scheme based on epitaxial thin films of the perovskite manganese oxide material $Nd_{0.67}Sr_{x0.33}MnO_3$, grown by pulsed laser deposition on buffered silicon substrates. The substrate and sensor materials are chosen to meet the conflicting requirements of radiation hardness, sensitivity, speed and linearity over a dynamic range of three orders of magnitude. The key challenge in the material development is the integration of the sensor material with Si. Si is required to withstand the free electron laser pulse impact and to achieve a readout speed three orders of magnitude faster than conventional cryoradiometers for compatibility with the Linac Coherent Light Source pulse rate. We discuss sensor material development and the photoresponse of prototype devices. This Linac Coherent Light Source total energy monitor represents the first practical application of manganite materials as bolometric sensors.

INTRODUCTION

The next generation X-ray sources based on free electron lasers (FEL) will generate ultra short coherent X-ray pulses through the process of self-amplified stimulated emission (SASE), whose peak brightness will exceed that of third generation synchrotrons by about ten orders of magnitude and their average brightness by three orders of magnitudes.[1,2] They will enable wide-ranging novel science from atomic and chemical dynamics studies[3] to single shot diffraction imaging.[4] Among the different FEL sources currently being built, the Linac Coherent Light Source (LCLS) at the Stanford Linear Accelerator Center (SLAC) will be the first to extend operation to X-ray energies of 0.8 to 8 keV. LCLS is expected to produce ~10^{12} X-rays per ~200 fs pulse with a repeat frequency of up to 120 Hz.[5] Due to the statistical character of the SASE process, the pulse-to-pulse variations in photon number will be significant,[2] and a crucial task will be to measure the energy of each pulse over the entire range of LCLS operating conditions.

Among the X-ray diagnostic instruments at LCLS, only a thermal detector is thought to be reliable, calibratable, and its response calculable in this new regime of ultra-short ultra-bright X-ray pulses where the response of other detectors will be non-linear or saturated, since most of the deposited laser energy will ultimately be converted into heat. A thermal total energy monitor can therefore be used during commissioning for X-ray FEL characterization and absolute calibration of other instruments, and during FEL operation for pulse intensity measurements at user end stations. For LCLS, this total energy monitor must be radiation hard in the X-ray band from 0.8 to 8 keV for energies up to ~2 mJ per pulse, must be sensitive down to the energy level of ~1 μJ of the spontaneous undulator radiation, absolutely calibratable to ≤ 10%, and must be compatible with an operating frequency of at least 10 Hz for commissioning, preferably 120 Hz.

In this letter, we demonstrate a bolometric detector scheme based on a hole doped rare earth manganite (also known as colossal magnetoresistive or 'CMR' manganite) sensor on a thin silicon substrate, for operation in the vicinity of its metal-insulator transition.[6,7] CMR manganites are promising sensor materials, since their metal insulator transition temperature is determined by their cationic composition and can thus be tailored for a desired operating temperature. This tunability of operating temperatures makes manganites uniquely suited of the present application which demands flexibility in the operating temperature to allow the final detector design to accommodate possible unknown effects related to matter-energy interactions under unprecedented conditions. This is our foremost reason for choosing manganites over other potential thermal detector candidates. Given the fact that manganite materials have been a focus of applied materials research in the past decade, it is also noteworthy that the present detector represents the first practical application of manganite thin films as bolometric detectors.

Si has emerged as the absorber/substrate material of choice, since its low atomic number makes it radiation hard[8], its absorption characteristics allow calibration with conventional optical lasers, and lattice engineering schemes have been demonstrated for the epitaxial growth of CMR manganites such as $La_{0.67}Sr_{0.33}MnO_3$ and $La_{0.67}Ba_{0.33}MnO_3$ on Si.[9,10,11]. A 500 μm Si chip will absorb >99.9% of the FEL energy at 8 keV while transmitting most of the spontaneous undulator radiation, and the subsequent increase in temperature can be measured as a change in resistance of the manganite sensor on the backside of the Si. The Si substrate also serves as the thermal link to the cold bath and sets the time constant of the pulse decay, and its high thermal diffusivity at low temperature makes it compatible with 120 Hz operation, .

For LCLS, sensor operation at lower temperatures is advantageous because of lower thermal noise and higher speed due to reduced phonon scattering. More specifically, 3D finite-

element simulations of the thermal response indicate that the signal in Si-based sensors operated below ≤200 K has decayed to ≤0.1% after ≤100 ms to allow operation at 10 Hz during commissioning. Operation at the eventual LCLS frequency of 120 Hz is possible for sensors with operating temperatures of ≤100 K.[12] However, many manganites with low metal-insulator transition temperature exhibit two-phase (metallic and insulating) coexistence accompanied by hysteresis in resistance vs. temperature and excess noise,[13] which precludes use in detector applications. For the desired temperature range, we identified $Nd_{1-x}Sr_xMnO_3$ (NSMO) with $x \approx 0.33$ as a suitable sensor material as it has no pronounced two-phase co-existence and negligible hysteresis.[6] High quality epitaxial films are required to maximize the temperature coefficient of resistance $TCR \equiv 1/R\ \partial R/\partial T$ and to minimize 1/f noise. We have therefore taken on the task to develop epitaxial thin NSMO films on (001) oriented Si substrates using pulsed laser deposition (PLD).

EXPERIMENTAL METHODS

The chemical reactivity of Si with manganites, lattice and thermal expansion mismatch, and the easy formation of amorphous SiO_2 preclude the growth of high quality films directly on bare Si and necessitate the use of buffer/template layers. Our initial efforts used Si substrates with a 100 Å buffer of high-quality epitaxial $SrTiO_3$ (STO) deposited by molecular beam epitaxy (MBE) at growth temperatures between ~ 400 and 550°C.[14] NSMO layers were then grown from a commercial bulk ceramic target by pulsed laser deposition (PLD), using a 248-nm KrF pulsed excimer laser (COMPEX 205) with 25 ns pulse duration at a repetition rate of 5 Hz and a fluence of ~2 J/cm^2. For a target-to-substrate distance of 8.5 cm this yielded typical deposition rates of ~0.3 Å per pulse. NSMO growth was optimized at a substrate temperature of ~780 °C

with 400 mTorr of O_2 during deposition, and a post deposition cooling rate of 1 ºC/min in ~400 Torr of O_2. Faster cooling rates encourage microcracks to develop in the NSMO due to differential thermal expansion coefficients (table 1). Structural analysis using a high resolution 4-circle X-ray diffractometer (Discover D8, Bruker AXS) shows that the films are phase-pure, epitaxial and in-plane aligned. The STO (002) out of plane peak gives a lattice parameter of 3.891 Å, close to the single crystal bulk value of 3.905 Å. The NSMO $(002)_{psuedocubic}$ out of plane peak gives a lattice parameter of 3.82 Å, which is smaller than the out-of-plane lattice constant 3.86 Å for bulk NSMO due to the lattice mismatch of STO and NSMO and the resulting tetragonal lattice distortion. Typical rocking angle widths for the STO and NSMO layers are 0.16° and 0.48° FWHM, respectively, indicating good crystallinity. The phi-scans show epitaxial in-plane alignment between the substrate and each of the different layers employed in the multilayer scheme. These NSMO films exhibit a metal-insulator transition around ~180 K and a maximum temperature coefficient of resistance TCR = 9.4 %/K at 143 K. The transition temperature is considerably suppressed and the peak resistivity is increased compared to strain-relaxed (thicker) NSMO films on $LaAlO_3$. This may be understood as due to the tensile strain from the lattice mismatch between NSMO and STO, which has been suggested to act as a Jahn-Teller type strain field, lifting the degeneracy of Mn^{3+} eg levels and introducing an energy barrier for the primary electron conduction mechanism by transfer between Mn^{3+} and Mn^{4+} ions.[15] In addition, the strained films are also prone to undesirable two-phase behavior.[16,17] Lattice mismatch strain is progressively relaxed as the films grow thicker.[18] However, in this case, growing thicker films is not an option because thermal strain due to different thermal expansion coefficients for Si and the oxide layers increases as the thickness is increased. Beyond a certain

thickness, which depends on the thermal kinetics of growth, the films are observed to develop microcracks.

We have therefore employed a second lattice matched template layer of $Bi_4Ti_3O_{12}$ (BTO) between the STO and NSMO layers. Figure 1 shows a typical 2θ-ω scan of a Si/STO/BTO/NSMO multilayer, showing the phase-pure, oriented layers. The BTO out of plane peaks corresponds to a lattice parameter of 32.62 Å, close to the single crystal bulk value of 32.81 Å. The NSMO $(002)_{psuedocubic}$ out of plane peak gives a lattice parameter of 3.85 Å. Note that this is very close to the lattice constant of ~3.86 Å for bulk NSMO, indicating that the lattice mismatch strain has been largely eliminated by the BTO template (table 1). Typical rocking angles for the BTO and NSMO are 0.13° and 0.23° FWHM, respectively, indicating improved NSMO crystallinity due to the BTO buffer. The metal-insulator transition temperature increases to ~200 K and the peak resistivity of the NSMO films is reduced by a factor of ~2, both closer to the optimized unstrained film values. Most importantly for bolometer applications, the maximum TCR is increased to ~13.5 %/K at ~175 K (figure 2), which is more than sufficient for the required sensitivity at LCLS.[7]

RESULTS AND DISCUSSION

The NSMO films have been photolithographically patterned into 2 × 2 mm^2 sensors, cooled inside a vacuum chamber by a low-vibration mechanical pulse-tube refrigerator, stabilized at the temperature of maximum TCR, and exposed to pulsed 532 nm radiation from a commercial Nd-YAG calibration laser (Coherent Minilite-I). Figure 3 shows the sensor response at pulse energy of 1.5 mJ when the 5 ns laser pulse is focused to ~1 mm FWHM and hits the backside of the Si substrate directly opposite the NSMO sensor. Note that in addition to the

expected thermal signal, there are two fast athermal transients in the response whose presence does not depend on the presence of the NSMO film, or the presence of piezoelectric buffers, or the application of a bias voltage. The first one (a) is consistent with electron-hole separation during diffusion due to their different mobilities (Dember effect).[19] The second one (b) is consistent with the diffusion of thermally generated majority carriers in the Si substrate from higher to lower temperature regions (transient Seebeck effect).[19] The sensor must be designed such that both transients have fully decayed before the thermal signal peaks, and can therefore not be designed for operation much below ~150 K where the thermal signal rise time is much faster[7,12]. For the prototype NSMO sensor discussed in figure 3, the transients have fully decayed after ~200 μs, so that the peak response at t = 250 μs is linear with incident pulse energy over the energy range of interest for LCLS (inset figure 3). Once the transients have decayed, both the peak signal and its integral can be used as measures of the incident energy, with the integral being somewhat less sensitive to beam jitter and variations X-ray absorption length over the LCLS energy range from 0.8 to 8 keV.[12] The observed signal decay times of ~10 ms are set by the diffusivity in Si and by the thermal coupling of the Si chip to the Cu chip holder and are three orders of magnitude faster than those of conventional cryoradiometers.[20]

CONCLUSION

These results demonstrate the feasibility of building a fast rare-earth manganite sensor based on a Si/STO/BTO/NSMO multilayer with high sensitivity and good linearity for total energy measurements at future free-electron X-ray laser facilities. Future work will focus on further reducing the 1/f noise to increase sensitivity at low FEL energies, and on reducing the response time for full compatibility with 120 Hz operation at LCLS. The latter will likely necessitate a suppression of the athermal transients through the appropriate choice of dopants in

the Si substrate, so that faster operation a lower temperatures is possible without interference of the transients with the thermal signal.

ACKNOWLEDGEMENTS

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. We acknowledge support from the undergraduate research grants to Sanjay Adhikari, Rajeh Mundle, David Cox and Anthony Davidson III from the Jess and Mildred Fisher College of Science and Mathematics, Towson University. Thanks are due to Jeff Klupt for help with laboratory instrumentation. RK acknowledges support from the Cottrell College Science Award from the Research Corporation through grant CC 6291.

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FIGURE LEGENDS

Figure 1: Diffraction (2θ/ω) scan of the (00l) peaks of a 800 Å $Nd_{0.67}Sr_{0.33}MnO_3$ (NSMO) film on Si, using a 100 Å $SrTiO_3$ buffer (STO) grown by MBE and a 340 Å $Bi_4Ti_3O_{12}$ buffer grown by PLD.

Figure 2: Metal-insulator transition in a 800 Å NSMO film of BTO-STO-buffered Si. The resistance peaks at ~201 K, and the TCR has a maximum value of ~12 %/K at ~176 K.

Figure 3: Laser induced response of a prototype total energy monitor. Note that the thermal signal (solid line) agrees with finite element simulations (dotted line), but is preceded by two non-thermal fast transients due to the Dember effect (a) and due to the transient thermoelectric effect (b). The inset shows the linearity of the thermal signal at t = 250 μs over the energy range of interest for LCLS.

TABLE

	Si (cubic)	**$SrTiO_3$** (cubic)	**$Bi_4Ti_3O_{12}$** (tetragonal)	**$Nd_{2/3}Sr_{1/3}MnO_3$** (pseudocubic)
Bulk thermal expansion coefficient α	2.49×10^{-6} K^{-1} *at 300K*	3.23×10^{-5} K^{-1} *300 to 2000K*		$\sim3\times10^{-5}$ K^{-1}
Bulk lattice parameters at 300K	a = 5.4309Å a/√2 = 3.840Å	a = 3.905Å	a = 3.861Å c = 32.81Å	a = 3.87Å
Si/STO/NSMO film: Lattice parameters Rocking angles		$a_\perp$ = 3.891Å FWHM = 0.16º		$a_\perp$ = 3.82Å FWHM = 0.48º
Si/STO/BTO/NSMO film: Lattice parameters Rocking angles			c = 32.62Å FWHM = 0.13º	$a_\perp$ = 3.85Å FWHM = 0.23º

Table 1: Summary of bulk and film characteristics.